

(RESEARCH ARTICLE)



Effect of CaCO₃ on the Formation corrosion and sedimentation in Iron pipe Drinking water in Kufra- Libya

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Abstract

Corrosion phenomena means to the deterioration of metallic materials due to chemical and electrochemical reactions, because these materials are always trying to reach a state of lower energy potential. Groundwater is vital and the sole resource in most of the studied region in the city of Alkufra. In this study, The main objectives of this research are to further broaden the mechanistic understanding of CO₂ corrosion of mild steel in the presence of high concentrations of calcium ions and evaluate the protectiveness of CaCO₃ scale against further corrosion. The values of these concentrations in the studied water samples indicate that they constitute a chemically suitable environment for corrosion and precipitation reactions, additionally, the pH values for the studied water samples ranged between (7.25 - 7.37), temperature values ranged from 35 °C to 37°C . (95.64 F0 to 98.6 F) respectively, it was found that the Aggressiveness coefficient ranged between (0.25 - (-1.3) and this indicates that the water in this region is highly corrosive, and the Ryzner coefficient ranges between (6.7 - 9.7), which indicates the water condition is the cause of corrosion. The Langelier coefficient index shows that the negative values are the cause of the increase in wear rate values from (-2.73 to -0.89) .Our results show that the presence of calcium carbonate and calcium sulfate as a scaling environment increases the corrosion rates for carbon steel.

Keywords: Water; Corrosion; Sedimentation; Al kufra; Ground

1 Introduction

Iron corrosion is one of the most complicated and costly problems facing drinking water utilities. A large number of parameters affect pipe corrosion, including water quality and composition, flow conditions, biological activity, and corrosion inhibitors. Corrosion studies in an attempt to provide the water industry with an updated understanding of factors that influence iron pipe corrosion. In particular, this paper reiterates conclusions of prior studies regarding the Langelier Index—despite its continued widespread use, the Langelier Index does not provide an effective means of controlling iron corrosion. A review of potential implications of upcoming regulations for iron corrosion is also included (1).

Precipitation of a thin layer of protective calcium carbonate (CaCO₃, presumably calcite) was the earliest proposed method for controlling iron corrosion. Many papers have been dedicated to extolling the virtues of calcite layers (2-3-9) and many others detail methods to achieve a perfect layer of protective calcite. However, few papers ever demonstrated a beneficial role for calcite in controlling iron corrosion .Calcium ions are usually present at high concentrations in brines produced with oil and gas. Such brines are often saturated with respect to calcium carbonate (CaCO₃). Consequently, precipitation of CaCO₃ as scale on the internal wall of the pipeline can readily occur due to changes in

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operational and environmental parameters as produced fluids are transferred from down hole to surface facilities. Despite its importance, there is minimal research in the literature addressing the effect of calcium ions, and specifically CaCO_3 scale, on the CO_2 corrosion mechanism. Several studies stressed the importance of “free CO_2 ”, which is the sum of carbonic acid and dissolved carbon dioxide. These studies found that the free CO_2 forms surface complexes on the iron oxide, increasing the solubility of ferrous iron and thus increasing iron concentration and corrosion rates⁽¹⁰⁾,⁽¹¹⁻¹³⁾.

The results of such studies are often contradictory and the corrosion mechanisms in the presence of alkaline earth cations, such as Ca^{2+} . There are few research studies in the literature that address the effect of Ca^{2+} ions on CO_2 corrosion⁽¹⁴⁾, have not been methodically characterized. There are studies claiming that corrosion rate is higher in the presence of Ca^{2+} ,⁽¹⁵⁻¹⁶⁾. Conversely, some researchers came up with the exact opposite conclusions⁽¹⁷⁻¹⁹⁾. There are claims that Ca^{2+} ions initiate pitting corrosion attack⁽¹⁹⁾; while another research study states that the presence of Ca^{2+} ions could postpone the occurrence of pitting on mild steel⁽²⁰⁾. Such discrepancies concerning the true effect of Ca^{2+} on CO_2 corrosion are often due to inadequate experimental procedures and setups which did not enable proper control of the solution chemistry. So this confusion found in the open literature prompted the development of different procedures and construction of a new experimental setup for elucidating the relevant issues surrounding CO_2 corrosion in the presence of Ca^{2+} ions.

Calcium carbonate (CaCO_3) is isostructural with FeCO_3 . Therefore, Ca^{2+} readily incorporates into the FeCO_3 structure and vice versa⁽²¹⁾. Therefore, the properties of FeCO_3 , both morphologically and chemically in the presence of Ca^{2+} , are subject to change. It has been established already that the morphology of FeCO_3 plays a significant role in the corrosion process⁽²²⁻²³⁾ the role of brine chemistry particularly in terms of the effect of individual ions on corrosion product layer formation, one of the important species in brines is Ca^{2+} , which can be present at high concentrations⁽²⁴⁾.” Ding, et al., reported the corrosion rate increased with an increase in the Ca^{+2} concentration⁽²⁵⁾.

The CO_2 corrosion mechanism of mild steel in the presence of high concentrations of Ca^{+2} suggests that the presence of Ca^{2+} in the solution and possible precipitation of CaCO_3 scale on the steel surface would influence the CO_2 corrosion mechanism. The solubility of CaCO_3 in water is about two orders of magnitude greater than the solubility of FeCO_3 . Therefore, substitution of Fe^{2+} by Ca^{2+} in the lattice of FeCO_3 can be hypothesized to alter the solubility of the mixed carbonate layers in comparison with pure FeCO_3 layers. In addition, compositional heterogeneity and morphological alteration are expected when Ca^{2+} incorporates into the FeCO_3 crystal structure. Mechanisms of CO_2 corrosion of mild steel and the characteristics of its corrosion products have been intensively studied and documented by different researchers⁽²⁶⁾. Such studies rely on the initial Ca^{2+} concentration rather than on the CaCO_3 saturation degree of the bulk solution as the core influential parameter. Iron carbonate (FeCO_3) is the main corrosion product in CO_2 corrosion. FeCO_3 is here referred to as a “corrosion product”, (Ca^{2+}) comes from the bulk solution and is then deposited on the steel surface⁽¹⁴⁾. This indicates that their constituent cations (Ca^{2+} and Fe^{2+}) can coexist in a substitutional mixed carbonate. CaCO_3 (mineralogical name: calcite) and FeCO_3 (siderite) are isostructural with a hexagonal unit cell, designated with the formula $\text{Fe}_x\text{Ca}_y\text{CO}_3$ ⁽³¹⁾, partial dissolution and/or destruction of this protective layer can lead to localized corrosion⁽³²⁾. With respect to CaCO_3 , precipitation of CaCO_3 is inevitable due to its fast kinetics; particularly at elevated temperatures. , the solution tends to reach an equilibrium state with a saturation with respect to CaCO_3 approaching unity. This can lead to significant changes in water chemistry (different pH, $[\text{Ca}^{2+}]$, from the initial conditions (to the final conditions). In that general corrosion was predominant other than pitting corrosion.

Ions (Ca^{2+} , CO_3^{2-}) plays a crucial role in the calculation of carbonate saturation in the bulk solution. For ideal solutions, the activity of ions is equal to its concentration. Activity is referred to as the effective concentration of an ion in a non-ideal solution. However, in the case of a non-ideal solution, the activity of a particular ion can be much lower or higher than its concentration⁽³³⁾. Therefore, ignoring the non-ideality of the solution can lead to miscalculations of CaCO_3 and FeCO_3 saturation degrees. This is especially true for studies focused on corrosion/scale interactions, where authors typically do not take non-ideality into consideration and consequently report incorrect values for the carbonates saturation degree⁽³⁴⁻³⁵⁾. Problems caused by water are plankton and chemical classes that lead to precipitation in the form of salts, crusts and chemical products. apart from this, depending on its quality and specifications, may be an aggressive environment for most of the materials, especially mineral ones, which makes it a major cause of the occurrence of corrosion and damage to various metals and alloys by chemical products dissolves in this medium and works to change its quality, physical appearance and quantity⁽³⁶⁾. A general phenomenon in our lives, it is widespread everywhere and in all environments, whether in the atmosphere, corrosion of wells, filters and pumps is a serious phenomenon that plays a major role in the rate of corrosion of metals^(37,38).

On the other hand. In general both iron concentration and the rate of corrosion increase with time when a pipe is first exposed to water, but then gradually both are reduced as the scale builds up high levels of chloride and sulfate in water can accelerate corrosiveness in the distribution networks⁽³⁹⁾. On the other hand, calcium carbonate can precipitate its

component, which is called cortical formation, which can add electrochemical corrosion reactions in the aqueous medium, especially with iron metals, as it gives balance with carbon dioxide and its cause of increased corrosion⁽⁴⁰⁻⁴¹⁾.

1.1 Study area location

In the city of Al Kufra .It was studied based on a group of groundwater wells at different. It is approximately in the middle of the northern corner of this basin

2 Material and methods

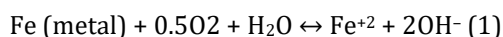
Water samples were collected from 8 different wells. The water samples were drawn from different depths this study we are doing some physical and chemical analysis on these Wells samples. (The pH value was measured using a pH device meter). The total amount of soluble salts (TDS). Immediately after collecting the samples using the Conductivity meter, bicarbonate was measured by titration using diluted standard sulfuric acid in the presence of an orange methyl guide, sulfate, and chloride. These measurements were determined according to the methods reported by⁽⁴²⁾.

3 Results and discussion

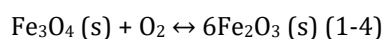
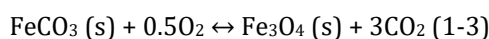
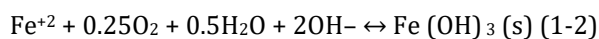
The results obtained were shown in Table (1) the rate of corrosion triples or quadruples as water temperature rises from 60°F to 140°F. Above 140°F the rate of corrosion doubles for every 20°F increase, thereby results in increase in temperature of the groundwater from 35°C to 37 °C and (95.64 F0 to 98.6 F) respectively, the role of temperature in corrosion is that all chemical reactions inside the pipes of groundwater are affected, stimulated, and their speed increases with a rise in temperature, which leads to an increase in the speed of corrosion of pipes. Also, the temperature affects the increase in the values of the solubility constant, the increase in the sedimentation speed, and thus the corrosion rate.

The effect of temperature on iron corrosion is often overlooked. Many parameters that influence corrosion can vary with temperature: dissolved oxygen (DO) solubility, solution properties (e.g. viscosity and ion mobility), ferrous iron oxidation rate, thermodynamic properties of iron scale (leading to formation of different phases or compounds), and biological activity.

Dissolved oxygen (DO) is an important electron acceptor in the corrosion of metallic iron



DO can also play a role in the oxidation of ferrous iron (Fe+2) or iron scales, for example:



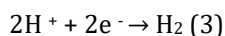
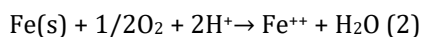
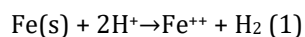
Thus, oxygen concentration can have varying effects on iron corrosion. As expected, the corrosion rate increases with increasing DO⁽⁴³⁾. However, effects on iron concentration and tuberculation may be mixed depending on the type of scale formed. Higher turbidity (a surrogate for iron concentration) was seen at lower oxygen saturation⁽⁴⁴⁾ but it is also reported that water free of DO will not tuberculate⁽⁴⁵⁾ DO is also responsible for the ability of buffering ions including phosphates, to inhibit corrosion⁽⁴⁶⁻⁴⁸⁾ For example, in water with DO < 1 mg L.

Kuch Mechanism In the absence of oxygen, it is possible for previously deposited ferric scale to act as an electron acceptor. This Kuch reaction produces ferrous iron and allows the corrosion reaction to continue even after DO is depleted :⁽⁴⁹⁻⁵⁰⁾



The pH plays an important role in determining the degree of corrosion, pH values for the studied water ranged between (7.25 - 7.38) PH. The presence of proteins constitutes half of the cathode reaction for the escalation of hydrogen gas and the increase in the rate of corrosion in the presence of a cathode reaction of half the oxygen reduction⁽¹¹⁾

If this medium is in direct contact with the iron metal, these reactions will take place



Water with pH values above 7.5 also can be corrosive when alkalinity is low⁽²³⁾ Role of pH. Weight loss is generally found to increase with increasing pH in the range 7 to 9.8 as is the degree of tuberculation.⁽⁵¹⁾ In contrast, by-product release was decreased at higher pH.7 Again, this is consistent with increased corrosion by-products being incorporated into the scale layer. However, one study found that both weight loss and iron concentration decreased as pH was raised from 8.5 to 9.2.⁽¹⁰⁾

The preferential dissolution of the ferrite phase (α -Fe) over Fe_3C in the corrosion process. It has been reported that the presence of Fe_3C increases corrosion rate through a galvanic effect and provides more cathodic sites for the hydrogen evolution reaction⁽⁵²⁾. The possible pathway for hydrogen evolution reactions in CO_2 aqueous environments can be described as follows:

At a higher temperature, the content of oxygen increases while the content of carbon in a protective layer decreases, which corresponds to the formation of divergent corrosion products at different temperatures. Iron carbonate (FeCO_3) was detected as the main component of the layer with highest protective performance.

Table 1 Results of the physical and chemical properties of groundwater samples in the city of Kufra

| NO.W | TC | SO ₄ ppm | HCO ₃ ppm | CO ₃ ppm | TDS ppm | PH | Ca ppm | Na ppm |
|------|------|------------------------|----------------------|------------------------|------------|------|--------|--------|
| P1 | 37 | 41.74 | 38.43 | 0 | 158 | 7.30 | 14.12 | 26.91 |
| P2 | 37 | 28.08 | 53.68 | 0 | 132 | 7.29 | 14 | 20.01 |
| P3 | 35.6 | 65.28 | 53.68 | 0 | 630 | 7.33 | 28. | 75.21 |
| P4 | 36.3 | 45.12 | 54 | 0 | 175 | 7.28 | 14 | 31.05 |
| P5 | 35 | 80.16 | 61 | 0 | 736 | 7.36 | 36 | 163.9 |
| P6 | 36.5 | 325 | 106.75 | 0 | 1288 | 7.25 | 76 | 296 |
| P7 | 36.6 | 285.6 | 285.6 | 0 | 641 | 7.37 | 74 | 169.97 |
| P8 | 37 | 54.72 | 54.72 | 0 | 272 | 7.32 | 26 | 40.94 |

Tests can determine if water is likely to be corrosive: the Langelier Saturation Index (LSI) and the Ryzner Stability Index (RSI) from the results obtained in Table (2). In order to use the LSI, a laboratory must measure pH, electrical conductivity, total dissolved solids, alkalinity, and total hardness. The LSI is typically negative or positive and only rarely zero. Negative values predict that the water is more likely to be corrosive. Potentially corrosive water typically has an LSI value -1 (mild) to -5 (severe). The Langelier Saturation Index is probably a good indication of corrosion or scaling potential of a well.

From the results obtained in Table (2). A parameter through which the aggressive index of the aqueous medium of well water is determined as it deals with the conditions of that water that have different exponent values. Hydrogen as well as basic and given by

$$\text{AL} = \text{pH} + \text{Log} (\text{ALK} * \text{Ca})$$

Where;

AL aggressiveness factor,

ALK total basal,

Ca Calcium ion concentration:

Where the values of indicators for the state of the studied water terms of corrosion and sedimentation showed that the aggressiveness factor ranges between (10.03 in P1) and (11.6 in P7). This indicates that the water is deposited by salts to cause highly corrosive. The Langelier index ⁽¹⁹⁾ so all the negative values Langelier Index but in p7 is positive. Is an indicator of the degree of saturation, sedimentation, calcium carbonate, or corrosion, and the molting of calcium carbonate?

Through the study of water corrosion, it became clear that the Ryzner coefficient in Table (2) ranges between (6.7 -9.7) is a clearer idea of the degree of water saturation and consequently, subject to precipitation or corrosion. These values are total above 6, and this, in fact, indicates that the water is corrosive. Medium corrosive environment. These results are consistent with and support the results obtained from Langelier⁽¹⁹⁾ coefficient, due to the fact that Both factors depend on the same variables in the calculations (pH, calcium, T.ALK, and temperature) Among the results obtained for estimating the aggressiveness factor, the values of the aggressiveness factor ranged between (0.25 in P7) and (-1.3) in P1).

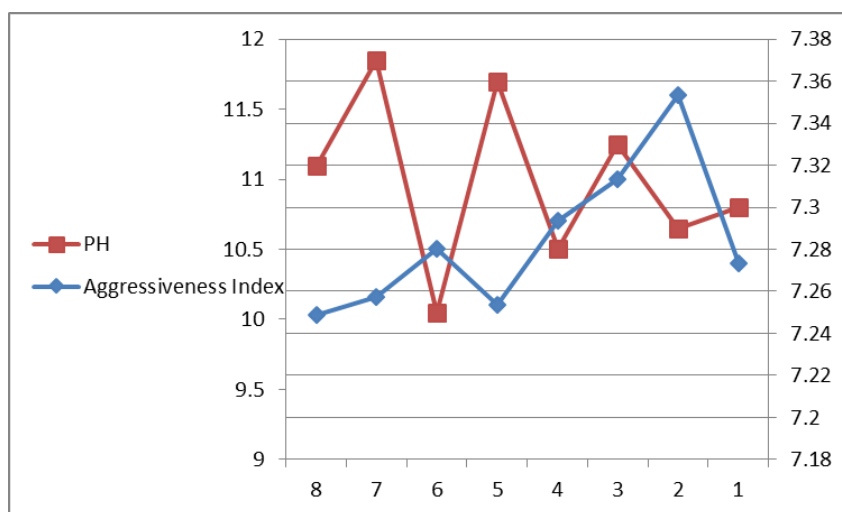


Figure 1 The PH and AL aggressiveness factor by $AL = pH + \text{Log} (ALK * Ca)$

The degree of corrosion of the aqueous medium, and these figures refer to the corrosion of the mean of the medium (water) and these results are consistent with. Those obtained from the previous two coefficients and support them, especially since which confirmed the corrosion of the aqueous medium and the absence of sedimentation on the water of the tubes.

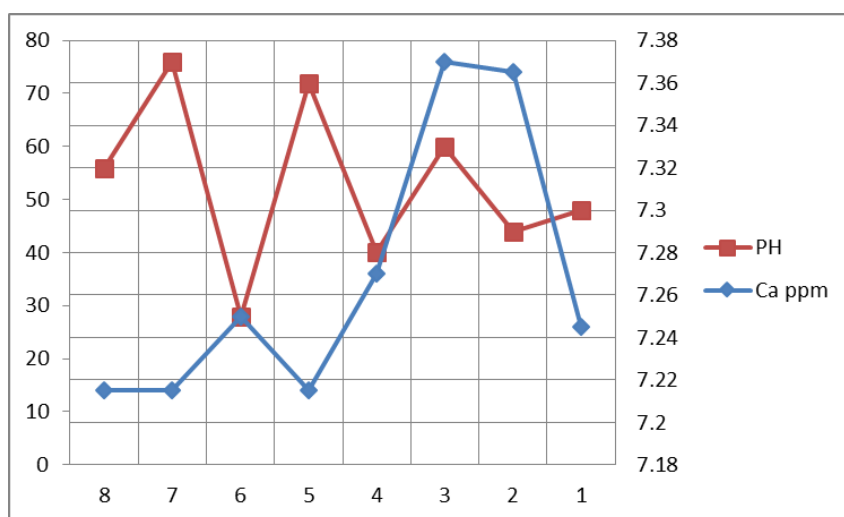


Figure 2 The relationship between PH and Ca^{+2} ppm

This compatibility is due to the fact that the aggressiveness coefficient depends on pH, calcium, T.ALK and temperature.

Table 2 A parameters through which the aggressive index, Ryzner ndex and Langelier index

| No.Well | Langelier Index | Aggressiveness Index | Ryzner Index |
|---------|-----------------|----------------------|--------------|
| P1 | -1.3 | 10.03 | 9.8 |
| P2 | -1.1 | 10.16 | 9.5 |
| P3 | 0.95- | 10.5 | 9.2 |
| P4 | -1.2 | 10.1 | 9.7 |
| P5 | -0.79 | 10.7 | 8.9 |
| P6 | -0.39 | 11 | 8 |
| P7 | 0.25 | 11.6 | 6.7 |
| P8 | -0.87 | 10.4 | 9.1 |

Langelier Index The indications for the LSI and the improved LSI by Carrier are based on the following values

-Water is undersaturated with respect to calcium carbonate. Undersaturated $LSI < 0$

-water has a tendency to remove existing calcium carbonate protective coatings in pipelines and equipment

$LSI = 0$

-Water is considered to be neutral. Neither scale-forming nor scale removing $LSI > 0$

Water is supersaturated with respect to calcium carbonate ($CaCO_3$) and scale forming may occur.

Table 3 LSI for level Corrosion

| Indication LSI (Carrier) | |
|--------------------------|------------------------------------------|
| 0,5->2,0- | Serious corrosion |
| 0>0,5- | Slightly corrosion but non-scale forming |
| LSI = 0,0 | Balanced but pitting corrosion possible |
| 0,5>0,0 | Slightly scale forming and corrosive |
| 2>0,5 | Scale forming but non corrosive |

Table 4 Ryzner Index for level Corrosion

| Indication (Carrier 1965) | |
|---------------------------|---------------------------|
| 5,0 - 4,0 | Heavy scale |
| 6,0 - 5,0 | Light scale |
| 7,0 - 6,0 | Little scale or corrosion |
| 7,5 - 7,0 | Corrosion significant |
| 9,0 - 7,5 | Heavy corrosion |
| 9,0< | Corrosion intolerable |

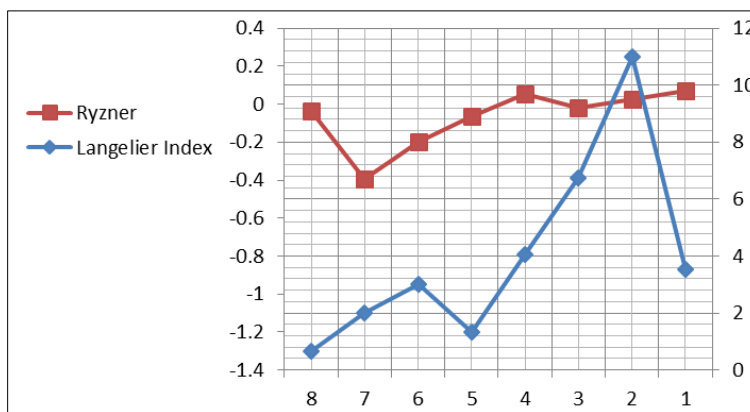


Figure 3 The relationship between Langelier index and Ryzner index

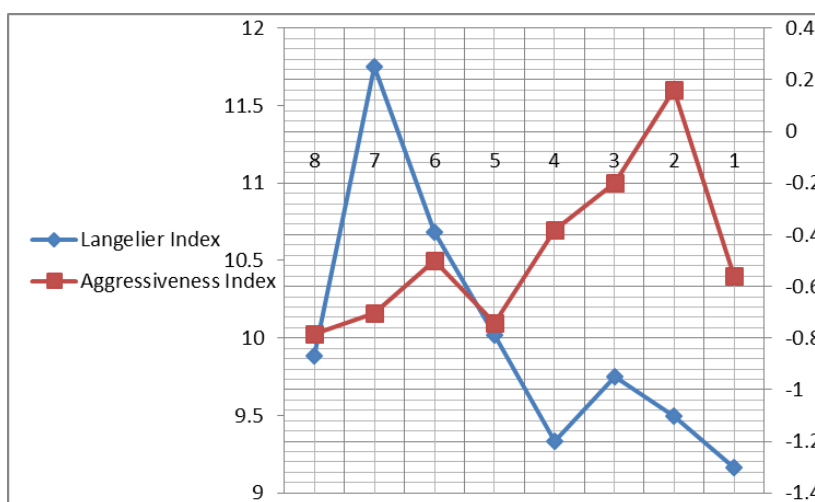


Figure 4 The relationship between Aggressive index and Langelier Index

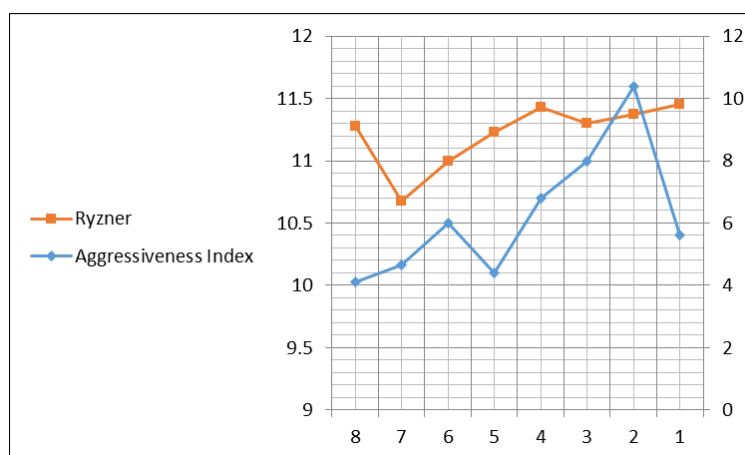


Figure 5 The relationship between Aggressive index and Ryzner index

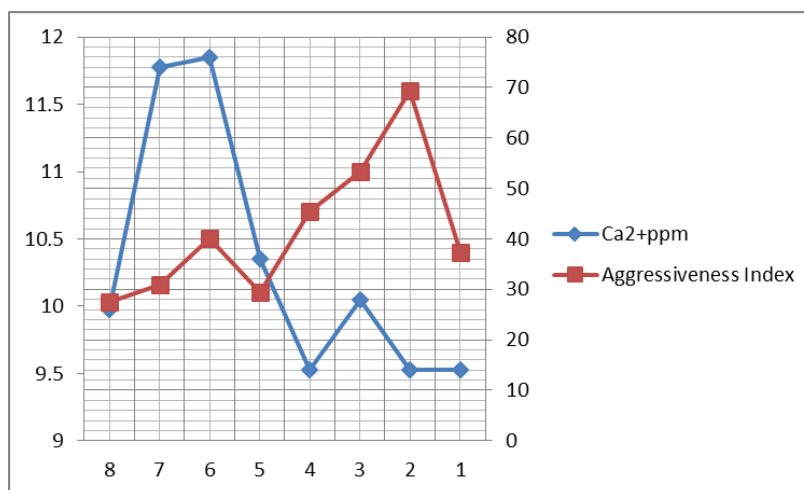
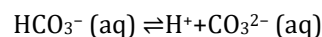
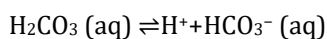
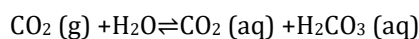


Figure 6 The relationship between Aggressiveness index and Ca²⁺ ppm

3.1 Impact of increasing temperature on CaCO₃ formation

While the levels of Ca²⁺ in natural waters are usually governed by regional geology (limestone, dolomite, etc.), the carbonate system is more complex. The pKa values indicate that CO₃²⁻ becomes the dominant form of inorganic carbon at pH 10.3



The ion activity product, Q, for CaCO₃ is the product of the Ca²⁺ and CO₃²⁻ activities as shown in the equation below.

$$Q = \{\text{Ca}^{2+}\} \{\text{CO}_3^{2-}\}$$

When $Q < K_{sp}$, the solution is undersaturated, precipitation is not thermodynamically favored, and solid CaCO₃ can dissolve. However, when $Q > K_{sp}$, CaCO₃ is supersaturated and can precipitate. Precipitation starts with nucleation

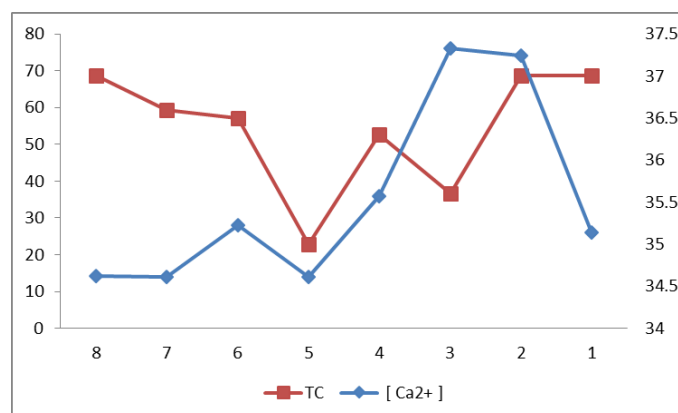


Figure 7 Effect the temperature on CaCO₃ formation

4 Conclusion

All of the treatments used and the tests that were studied in the assessment of the degree of corrosion and sedimentation, some chemical compounds confirmed that corrosion in the study areas corrosion ranges from the degree of corrosion to medium to severe corrosion. the Aggressiveness coefficient highly degree and this indicates that the

water in this region is highly corrosion, and the Ryzner coefficient, which indicates that the water condition is the cause of corrosion. The presence of high concentrations of chloride ion that will attack the steels, thereby creating dissolved products in the aqueous medium. It became clear through studying the physical and chemical properties of groundwater that it is corrosive and has a negative impact on mineral equipment and devices and therefore leads to a high rate of economic losses. Groundwater in the area should be monitored, programs established for treatment and control of concentrations and values that improve quality. Control its corrosive property. These results indicate the unsaturation of the wells studied, and this fact indicates the erosive and aggressive process of groundwater in the area corrosion. Our results show that the presence of calcium carbonate and calcium sulfate as a scaling environment increases the corrosion rates for aluminum alloys and carbon steel; however, the same environments do not affect the corrosion behavior of stainless steel. Our results contribute to the understanding and study of a topic we believe is in its infancy: the simultaneous scaling and corrosion phenomena on materials employed in the oil and gas industry. We hope that the results presented herein draw attention to

- The lack of research in the internal corrosion of steel and aluminum pipelines.
- The need to evaluate these materials for tubular manufacturing for the oil and gas industry.

Compliance with ethical standards

Disclosure of conflict of interest

No conflict of interest to be disclosed.

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